


TRANSMITTAL LETTER OF THE UNITED STATES DESIGNATED/ELECTED OFFICE (DO/EO/US) CONCERNING A FILING UNDER 35 U.S.C. 371		Attorney Docket No. <b>2001-1019</b>
		U.S. Application No. <b>10/070317</b>
INTERNATIONAL APPLN. NO. <b>PCT/NL00/00626</b>	INTERNATIONAL FILING DATE <b>6 September 2000</b>	PRIORITY DATE CLAIMED <b>6 September 1999</b>
TITLE OF INVENTION: <b>REDUCTION OF N<sub>2</sub>O EMISSIONS</b>		
APPLICANT(S) FOR DE/EO/US: <b>MICHAEL JOHANNES FRANCISCUS MARIA VERHAAK</b>		
Applicant herewith submits to the United States Designated Elected Office (DO/EO/US) the following items and other information:		
<ol style="list-style-type: none"> <li>1. <input checked="" type="checkbox"/> This is a FIRST submission of items concerning a filing under 35 U.S.C. 371.</li> <li>2. <input type="checkbox"/> This is a SECOND or SUBSEQUENT submission of items concerning a filing under 35 U.S.C. 371.</li> <li>3. <input checked="" type="checkbox"/> This is an express request to begin national examination procedures (35 U.S.C. 371(f)). The submission must include items (5), (6), (9) and (21) indicated below.</li> <li>4. <input checked="" type="checkbox"/> The US has been elected by the expiration of 19 months from the priority date (Article 31).</li> <li>5. <input checked="" type="checkbox"/> A copy of the International Application as filed (35 U.S.C. 371 (c)(2)) <ol style="list-style-type: none"> <li>a. <input checked="" type="checkbox"/> is attached hereto (required only if not communicated by the International Bureau)</li> <li>b. <input type="checkbox"/> has been communicated by the International Bureau. See attached PCT/IB/308.</li> <li>c. <input type="checkbox"/> is not required, as the application was filed in the United States Receiving Office (RO/US).</li> </ol> </li> <li>6. <input type="checkbox"/> An English language translation of the International Application as filed (35 U.S.C. 371 (c)(2)) <ol style="list-style-type: none"> <li>a. <input type="checkbox"/> is attached hereto.</li> <li>b. <input type="checkbox"/> has been previously submitted under 35 U.S.C. 154(d)(4).</li> </ol> </li> <li>7. <input type="checkbox"/> Amendments to the claims of the International Application under PCT Article 19 (35 U.S.C. 371 (c)(3)) <ol style="list-style-type: none"> <li>a. <input type="checkbox"/> are attached hereto (required only if not communicated by the International Bureau).</li> <li>b. <input type="checkbox"/> have been communicated by the International Bureau.</li> <li>c. <input type="checkbox"/> have not been made, however, the time limit for making such amendments has NOT expired.</li> <li>d. <input type="checkbox"/> have not been made and will not be made.</li> </ol> </li> <li>8. <input type="checkbox"/> An English language translation of the amendments to the claims under PCT Article 19 (35 U.S.C. 371 (c)(3)).</li> <li>9. <input checked="" type="checkbox"/> An oath or declaration of the inventor(s) (35 U.S.C. 371(c)(4)).</li> <li>10. <input type="checkbox"/> An English language translation of the annexes of the International Preliminary Examination Report under PCT Article 36 (35 U.S.C. 371(c)(5)).</li> </ol>		
Items 11 to 20 below concern document(s) or information included:		
<ol style="list-style-type: none"> <li>11. <input checked="" type="checkbox"/> Information Disclosure Statement (IDS) w/PTO-1449 - <input type="checkbox"/> Copy of IDS citations</li> <li>12. <input checked="" type="checkbox"/> Assignment Papers (cover sheet &amp; document(s))</li> <li>13. <input checked="" type="checkbox"/> A FIRST Preliminary Amendment.</li> <li>14. <input type="checkbox"/> A SECOND or SUBSEQUENT Preliminary Amendment.</li> <li>15. <input type="checkbox"/> A substitute specification.</li> <li>16. <input type="checkbox"/> A change of power of attorney and/or address letter.</li> <li>17. <input type="checkbox"/> A computer-readable form of the sequence listing in accordance with PCT Rule</li> <li>18. <input type="checkbox"/> A second copy of the published international application under 35 U.S.C. 154(d)(4).</li> <li>19. <input type="checkbox"/> A second copy of the English language translation of the international application (35 U.S.C. 154(d)(4)).</li> <li>20. <input checked="" type="checkbox"/> Other items or information: Application Data Sheet, Abstract on a separate sheet, International Preliminary Examination Report (PCT/IPEA/409), International Search Report (PCT/ISA/210)</li> </ol>		

U.S. APPLICATION NO. <b>10707031</b>		INTERNATIONAL APPLN. NO. PCT/NL00/00626		ATTORNEY DOCKET NO. 2001-1019	
21. <input checked="" type="checkbox"/> The following fees are submitted:  BASIC NATIONAL FEE (37 CFR 1.492 (a) (1)-(5):  Neither international preliminary examination fee nor international search fee paid to USPTO and international Search Report not prepared by the EPO or JPO .....\$1040.00  International preliminary examination fee not paid to USPTO but International Search Report prepared by the EPO or JPO.....\$890.00  International preliminary examination fee not paid to USPTO but International search fee paid to USPTO .....\$740.00  International preliminary examination fee paid to USPTO but all claims did not satisfy provision of PCT Article 33 (1)-(4) .....\$710.00  International preliminary examination fee paid to USPTO and all claims satisfied provision of PCT Article 33 (1)-(4).....\$100.00  <b>ENTER APPROPRIATE BASIC FEE AMOUNT</b>				CALCULATIONS PTO USE ONLY	
				\$ 890.00	
Surcharge of \$130.00 for furnishing the oath or declaration than <input type="checkbox"/> 20- <input type="checkbox"/> 30 Months from the earliest claimed priority date (37 CFR 1.492(e))				\$	
CLAIMS	NUMBER FILED	NUMBER EXTRA	RATE		
Total Claims	8 - 20 =	0	X \$18.00	\$	
Independent Claims	1 - 3 =	0	X \$84.00	\$	
MULTIPLE DEPEND CLAIM(S) (if applicable)			+ \$280.00	\$	
<b>TOTAL OF ABOVE CALCULATION -</b>				<b>\$ 890.00</b>	
<input type="checkbox"/> Applicant claims small entity status. See 37 CFR 1.27. The fees indicated above are reduced by 1/2.				\$	
<b>SUBTOTAL =</b>				<b>\$ 890.00</b>	
Processing fee of \$130.00 for furnishing the English translation later than <input type="checkbox"/> 20 <input type="checkbox"/> 30 months from the earliest claimed priority date (37 CFR 1.492Z(f)).				\$	
<b>TOTAL NATIONAL FEE =</b>				<b>\$ 890.00</b>	
Fee for recording the enclosed assigned (37 CFR 1.21(h)). The assignment must be accompanied by an appropriate cover sheet (37 CFR 3.28, 3.31) \$40.00 per property +				\$ 40.00	
<b>TOTAL FEES ENCLOSED -</b>				<b>\$ 930.00</b>	
				Amount to be refunded:	\$
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<input checked="" type="checkbox"/> A Check in the amount of <b>\$930.00</b> to cover all fees is attached.  <input type="checkbox"/> The Commissioner is hereby authorized to charge indicated fees and credit any overpayments to Deposit account No. 25-0120 in the name of Young & Thompson, as described below. A duplicate copy of this sheet is enclosed.  <input checked="" type="checkbox"/> The Commissioner is hereby authorized in this, concurrent, and future replies, to charge payment or credit any overpayment to Deposit Account No. 25-0120 for any additional fee required under 37 C.F.R. §§ 1.16 or 1.17.					
SEND ALL CORRESPONDENCE TO: 745 South 23 <sup>rd</sup> Street Arlington, VA 22202 Telephone (703) 521-2297 Y&T Customer No. 000466			 <b>00466</b> <small>PATENT - TRADEMARK OFFICE</small>		
BC/lmt Date: <b>5 MARCH 2002</b>			SIGNATURE <u><i>Benoit Castel</i></u> Benoit Castel NAME 35,041 REGISTRATION NO.		

10/070317

JC19 Rec'd PCT/PTO 05 MAR 2002

PATENT  
2001-1019

**IN THE U.S. PATENT AND TRADEMARK OFFICE**

In re application of: Michael Johannes Franciscus Maria VERHAAK

Appl. No.: **NEW NATIONAL PHASE  
APPLICATION IN THE  
UNITED STATES**

Filed: March 5, 2002

For: REDUCTION OF N<sub>2</sub>O EMISSIONS

**PRELIMINARY AMENDMENT**

Assistant Commissioner for Patents  
Washington, DC 20231

March 5, 2002

Sir:

The following preliminary amendments and remarks  
are respectfully submitted in connection with the above-  
identified application.

**IN THE ABSTRACT OF THE DISCLOSURE:**

Please insert the Abstract of the Disclosure  
attached on a separate sheet attached hereto.

**IN THE CLAIMS:**

Please amend the claims as follows:

--3. (amended) Method according to Claim 2,  
characterized in that the precious metal comprises ruthenium,  
rhodium, palladium, gold or platinum, or a combination of two  
or more of these metals.--

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--5. (amended) Method according to Claim 4, characterized in that the concentration of the reducing agent is set in such a way that the hydrocarbon/N<sub>2</sub>O molar ratio is in the range from 0.2 to 20, preferably in the range from 0.5 to 5.--

REMARKS

Claims 1-8 are pending in the present application.

Entry of the above amendments is earnestly solicited. An early and favorable first action on the merits is earnestly requested.

Should there be any matters that need to be resolved in the present application, the Examiner is respectfully requested to contact the undersigned at the telephone number listed below.

Attached hereto is a marked-up version of the changes made to the claims by the current amendment. The attached page is captioned "VERSION WITH MARKINGS TO SHOW CHANGES MADE."

The Commissioner is hereby authorized in this, concurrent, and future replies, to charge payment or credit any overpayment to Deposit Account No. 25-0120 for any additional fees required under 37 C.F.R. § 1.16 or under 37 C.F.R. § 1.17.

Respectfully submitted,

YOUNG & THOMPSON

*Benoit Castel*

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Attachments

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ABSTRACT OF THE DISCLOSURE

A method for the selective catalyst reduction of nitrous oxide ( $N_2O$ ) in the presence of a solid catalyst, with the addition of a saturated hydrocarbon as reducing agent. The catalyst used is a promoted, iron-containing zeolite. As a result, it is possible to increase the conversion of ( $N_2O$ ) compared to unpromoted iron-containing zeolite catalysts. The promoted catalyst is active at temperatures below  $350^{\circ}C$ .

Furthermore, it has been found that the iron-containing zeolite catalysts which are promoted with precious metal also give low emissions of CO and residual hydrocarbons. The catalyst is also very active under high process pressures and in the presence of sulphur.

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VERSION WITH MARKINGS TO SHOW CHANGES MADE

IN THE CLAIMS:

The claims have been amended as follows:

--3. (amended) Method according to Claims ~~1 and~~ 2, characterized in that the precious metal comprises ruthenium, rhodium, palladium, gold or platinum, or a combination of two or more of these metals.--

--5. (amended) Method according to Claims ~~1 and~~ 4, characterized in that the concentration of the reducing agent is set in such a way that the hydrocarbon/N<sub>2</sub>O molar ratio is in the range from 0.2 to 20, preferably in the range from 0.5 to 5.--

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### Reduction of N<sub>2</sub>O emissions

#### **Background**

5 Nitrous oxide (dinitrogen oxide, N<sub>2</sub>O) makes a substantial contribution to the greenhouse effect. The global warming potential (the extent to which a molecule contributes to the greenhouse effect compared to one molecule of CO<sub>2</sub>) of N<sub>2</sub>O is approx. 310. For a number of years, the policy of reducing emissions of greenhouse gases has been developed. The present invention can make a significant contribution to  
10 this policy. Various significant sources of N<sub>2</sub>O emissions have been identified: agriculture, industrial production of nylon precursors (adipic acid and caprolactam), the production of nitric acid and motor vehicles fitted with a three-way catalyst.

In principle, various catalytic and non-catalytic techniques can be employed in order to render nitrous oxide harmless. Various catalysts are known for the direct  
15 catalytic decomposition of N<sub>2</sub>O to N<sub>2</sub> and O<sub>2</sub> (cf. the literature summary provided by Kapteijn et al., Appl. Catal. B9 (1996), pp 25-64 and US-A-5,171,553). However, this reaction is hampered to a considerable extent by the presence of oxygen and water, which are to be found in the off-gases from virtually all the N<sub>2</sub>O sources listed above. Selective catalytic reduction is a promising alternative. Various catalysts for the  
20 conversion of N<sub>2</sub>O with the aid of olefins (C<sub>n</sub>H<sub>2n</sub>), alcohols or ammonia have been studied in the literature (recently: Mauzevin et al. Appl. Catal. B23 (1999) L79-L82 and Pophal et al. Appl Catal. B16 (1998) pp. 177-186 and the literature cited therein). Catalysts employed are often zeolites which have been substituted with a transition metal, such as iron, cobalt or copper.

25 For both technical and economic reasons, the addition of saturated hydrocarbons (C<sub>n</sub>H<sub>2n+2</sub>) would be preferable to the abovementioned reducing agents. Natural gas (CH<sub>4</sub>) and LPG (mixture of C<sub>3</sub>H<sub>8</sub> and C<sub>4</sub>H<sub>10</sub>) are particularly attractive in this context. It is important that the formation and emission of carbon monoxide (CO) and emission of unreacted hydrocarbons be minimized.

30 The present invention relates to a catalyst which enables N<sub>2</sub>O to be converted into nitrogen at a relatively low temperature and which allows very low emissions of CO and hydrocarbons to be achieved.

#### **Prior art**

35 International Patent Application WO 9949954 has described a method for the catalytic reduction of N<sub>2</sub>O in the presence of a zeolite with the addition of a reducing agent, the reducing agent used being a saturated hydrocarbon, such as methane (CH<sub>4</sub>), propane (C<sub>3</sub>H<sub>8</sub>), LPG (C<sub>3</sub>H<sub>8</sub>/C<sub>4</sub>H<sub>10</sub>), or a combination of these reducing agents. It has

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been found that complete conversion of  $N_2O$  can be achieved at reaction temperatures of  $400^\circ C$  or lower, even with very low concentrations of the reducing agent and in the presence of water vapour, oxygen and sulphur dioxide. The catalyst involves a specially prepared iron-substituted zeolite.

5 Japanese patent publications JP 05103953 and JP 07213864 describe the removal of  $N_2O$  in the presence of, respectively, methane and propane with the aid of (inter alia) and iron-zeolite catalyst. However, these methods take no account of the emission of CO and unreacted hydrocarbons.

To prevent undesirable emission of CO and residual hydrocarbons,  
10 Japanese patent publication JP 09000884 describes the mechanical mixing of an iron-zeolite catalyst with a supported platinum or palladium catalyst. The  $N_2O$  conversion is around 60% at  $450^\circ C$  in a gas which contains oxygen and water. The conversion of  $N_2O$  achieved with the mechanically mixed catalyst is significantly worse than with the iron-zeolite catalyst alone.

15 JP 05103953, JP 07213864 and JP 09000884 all lack information about the effectiveness of the invention under pressure and/or in the presence of sulphur compounds. This is essential for use in the production of nitric acid and caprolactam, respectively.

## 20 **Discovery of a novel catalyst**

One object of the present invention is to provide a method for the removal of  $N_2O$  from industrial gas streams which contain  $O_2$ ,  $H_2O$ ,  $NO_x$  and possibly sulphur and which may be at elevated pressure. A further object of the present invention is to bring about the abovementioned removal of  $N_2O$  by the addition of saturated  
25 hydrocarbons at a reaction temperature of lower than  $400^\circ C$ , with very low emissions of CO and unreacted hydrocarbons.

To this end, the method according to the invention is characterized in that the catalyst used is a promoted, iron-containing zeolite. The zeolite catalyst is preferably promoted with a precious metal.

30 The use of an iron-containing zeolite catalyst which has been promoted preferably with precious metal (Rh, Pd, Ru, Pt, Au, etc.) in the SCR of  $N_2O$  with the aid of saturated hydrocarbons ( $C_nH_{2n+2}$ ) has surprisingly led to increased conversion of  $N_2O$  compared to an unpromoted iron-containing zeolite catalyst. The promoted catalyst is active at temperatures of below  $350^\circ C$ .

35 Furthermore, it has been found that the iron-containing zeolite catalyst which has been promoted with precious metal reduces the emissions of CO and residual

hydrocarbons very considerably at the operating temperature compared to an unpromoted iron-containing zeolite catalyst.

In addition, it has been found that the catalyst according to the invention is also active in the removal of nitrogen oxides ( $\text{NO} + \text{NO}_2 = \text{NO}_x$ ). This is important in view of the fact that  $\text{NO}_x$  are also released from the various sources of  $\text{N}_2\text{O}$ .

It has also been found that the catalyst described is also very active in the desired conversion at elevated process pressure, at which it is even more active than at atmospheric pressure. This is an important fact with regard to its application in the nitric acid industry.

Finally, it has been found that the catalyst described is active if sulphur is present in the feed. This is an important fact in connection with its application in the nylon industry.

The invention will be explained in more detail with reference to the following examples together with the associated figures, in which:

Figure 1 shows the degree of conversion of  $\text{N}_2\text{O}$  as a function of temperature for four iron-containing zeolite catalysts promoted with precious metal (A, D, E and F). For comparison purposes, the  $\text{N}_2\text{O}$  conversion achieved by an unpromoted iron-containing zeolite catalyst (X) is also shown. The test conditions are described in Table 2 ( $\text{SV} = 19,500 \text{ h}^{-1}$ , pressure = 3 bara,  $\text{C}_3\text{H}_8$  concentration = 1900 ppmv).

Figure 2 shows the CO emissions which occur during the conversion of  $\text{N}_2\text{O}$  using the same catalysts and under the same conditions as in Figure 1.

#### I. Preparation of the catalysts

The catalysts according to the present invention are produced by adding zeolite Na-ZSM-5 or  $\text{NH}_4\text{-ZSM-5}$  to a solution of  $(\text{NH}_4)_2\text{Fe}(\text{SO}_4)_2 \cdot 6\text{H}_2\text{O}$  (Mohr's salt). After they have been combined, ion exchange is carried out for 8 hours at  $80^\circ\text{C}$ . The resulting suspension is filtered, the solid material is washed, is dried at  $80^\circ\text{C}$  and is calcined at  $550^\circ\text{C}$ .

The Fe-ZSM-5 base material obtained in this way is then impregnated with a volume of a solution of a precious metal precursor in demineralized water which is such that the pores of the base material are precisely filled (so-called incipient wetness impregnation). The concentration of the precious metal precursor is set in such a manner that the desired quantity of precious metal on the base material is obtained. Finally, the catalyst is dried at  $80^\circ\text{C}$  and calcined at  $550^\circ\text{C}$ . The catalyst powder obtained is pressed into a tablet, is ground and is screened.

The following catalysts are used in the examples:

Table 1

Catalyst	Base	Precious metal
A	Fe-ZSM-5 from Mohr's salt	0.05% Pd
B	Fe-ZSM-5 from Mohr's salt	0.1% Pd
C	Fe-ZSM-5 from Mohr's salt	0.3% Pd
D	Fe-ZSM-5 from Mohr's salt	0.3% Rh
E	Fe-ZSM-5 from Mohr's salt	0.3% Ru
F	Fe-ZSM-5 from Mohr's salt	0.3% Au
X	Fe-ZSM-5 from Mohr's salt	none

## II. Test apparatus

The conversion of  $N_2O$  by means of SCR using propane and methane was studied in an automated flow arrangement. The gases  $N_2$ , air,  $N_2O$ ,  $C_3H_8$ ,  $CH_4$ ,  $NO$ ,  $NO_2$  are introduced by means of calibrated mass flow controllers (Brooks). Water is added via a Liquiflow controller and a controlled Evaporator Mixer (Bronkhorst). The gases emerging are analysed by means of a calibrated FTIR spectrophotometer (Elsag, Bailey, Hartmann & Brown, type MB 100). The catalyst is in a stainless steel reactor. The gases are passed through a preheating section before they come into contact with the catalyst. The temperature at the entry to and exit from the catalyst bed is measured using thermocouples. The mean of these two temperatures is shown in the results of the tests. The pressure in the test arrangement can be set at levels of between 1 and 5 bar absolute (bara).

The gas composition in the examples is representative for use of the catalyst according to the present invention in the nitric acid industry. The general test conditions are as follows:

Table 2

Weight of catalyst	6 – 15 g
Screening fraction	0.71 – 1.4 mm
Volume of catalyst	10 – 23 ml
Total gas flow rate	5 – 7.5 l/min (STP)
Space velocity	13,000 – 45,000 h <sup>-1</sup>
Total pressure	1 – 5 bara
Temperature of catalyst	150 – 500 °C
O <sub>2</sub> concentration	2.5% v/v
H <sub>2</sub> O concentration	0.5% v/v
N <sub>2</sub> O concentration	1500 ppmv
C <sub>3</sub> H <sub>8</sub> concentration	1500 – 2500 ppmv
NO <sub>2</sub> concentration	100 ppmv
NO concentration	100 ppmv

5

### III. Influence of the addition of precious metal to iron-containing zeolite

Figure 1 shows the N<sub>2</sub>O conversion curves for the catalyst from Table 1 (test conditions as in Table 2, SV = 19,500 h<sup>-1</sup>, pressure = 3 bara, C<sub>3</sub>H<sub>8</sub> concentration = 1900 ppmv). The promoted catalyst (A,D,E,F) are more active than the unpromoted iron-containing zeolite catalyst X. This is evident from the shift of the N<sub>2</sub>O conversion curves towards a lower temperature compared to the unpromoted Fe-ZSM-5 catalyst X.

10

Figure 2 shows the CO concentrations as a function of temperature for the same catalysts under the same conditions. When using catalyst X and catalyst F, CO is

formed throughout the entire temperature range. The other catalysts are excellent at eliminating CO emissions: if catalyst E is used, the amount of CO at over 350°C is lower than 10 ppmv, and with catalysts A and D this level is already achieved from 300°C.

- 5 Comparative tests between catalyst A (containing 0.05% Pd) and catalysts B and C (respectively containing 0.1% and 0.3% Pd) reveal similar curves for both N<sub>2</sub>O conversion and CO formation. This means that a low concentration of precious metal is sufficient, reducing the cost of the catalyst.

- 10 Table 3 shows the concentrations of the compounds which leave the reactor at a mean temperature of 350°C for a number of catalysts (test conditions as in Table 2, SV = 19,500 h<sup>-1</sup>, pressure = 3 bara, C<sub>3</sub>H<sub>8</sub> concentration = 1900 ppmv).

Table 3

Catalyst	N <sub>2</sub> O (ppmv)	C <sub>3</sub> H <sub>8</sub> (ppmv)	CO (ppmv)	NO <sub>x</sub> <sup>a</sup> (ppmv)
C	70	62	3	85
D	55	33	1	85
E	130	75	37	82
F	42	70	2180	45
X	123	167	2267	46

<sup>a</sup>NO<sub>x</sub> = NO + NO<sub>2</sub>

- 15 For all the catalysts, the N<sub>2</sub>O and C<sub>3</sub>H<sub>8</sub> conversion levels are higher than 90% at 350°C. For the catalysts which contain Pd and Rh, the CO emission is negligible. Another surprise is that the catalysts also remove from 60 to 75% of the NO<sub>x</sub>.

#### IV. Activity of the promoted iron-containing zeolite catalysts at increased process pressure and space velocity

- 20 Increasing the process pressure has a beneficial affect on the activity of the catalysts. Table 4 shows the concentrations of a number of components of the emerging gas, at 1, 3 and 5 bara and at a reaction temperature of 350°C (test conditions as in Table 2, catalyst A, SV = 19,500h<sup>-1</sup>, C<sub>3</sub>H<sub>8</sub> concentration = 1900 ppmv). The N<sub>2</sub>O

conversion level remains greater than 90%, while propane slippage, CO and NO<sub>x</sub> emissions fall as the process pressure rises.

**Table 4**

Pressure (bara)	N <sub>2</sub> O (ppmv)	C <sub>3</sub> H <sub>8</sub> (ppmv)	CO (ppmv)	NO <sub>x</sub> <sup>a</sup> (ppmv)
1	95	200	14	137
3	128	124	3	135
5	100	54	2	122

<sup>a</sup>NO<sub>x</sub> = NO + NO<sub>2</sub>

5

At a process pressure of 4 bara and under conditions as in Table 2 (catalyst A, SV = 13,000 h<sup>-1</sup>), increased activity was measured when the propane concentration was increased from 1500 to 2000 ppmv. Further increasing the propane/N<sub>2</sub>O ratio had no further positive effect on the conversion of N<sub>2</sub>O.

10

Experiments under conditions as in Table 2 (catalyst A, pressure = 4 bara, C<sub>3</sub>H<sub>8</sub> concentration = 1900 ppmv) indicate that the space velocity can be increased from 13,000 to 45,000 h<sup>-1</sup> without the activity of the catalyst being adversely affected.

15

The stability of the catalyst is tested for 50 hours in the conditions as described in Table 2. No deterioration in the activity was detected.

#### V. N<sub>2</sub>O conversion in different gas compositions

Table 5 demonstrates that the catalyst according to the present invention also functions well with higher water concentrations and higher oxygen concentrations.

20

The table describes experiments in different gas compositions (conditions as in Table 2, catalyst B, SV = 13,000 h<sup>-1</sup>, no NO and NO<sub>2</sub> present)

Table 5

Feed				Reaction T = 350°C	
N <sub>2</sub> O (ppmv)	C <sub>3</sub> H <sub>8</sub> (ppmv)	O <sub>2</sub> (% v/v)	H <sub>2</sub> O (% v/v)	N <sub>2</sub> O- conversion	CO (ppmv)
500	1000	6	0.5	97%	3
500	1000	3	0.5	97%	2
500	1000	2	0.5	84%	3
1000	1000	2	0.5	90%	2
1000	500	2	0.5	82%	2
500	1000	6	2	94%	2
1000 <sup>a</sup>	1000	2	0.5	73% <sup>a</sup>	1 <sup>a</sup>

<sup>a</sup> In the presence of 250 ppmv NO

5

The catalyst X was tested in the presence of SO<sub>2</sub> (500 ppmv N<sub>2</sub>O, 500 ppmv C<sub>3</sub>H<sub>8</sub>, 6% H<sub>2</sub>O, 500 ppmv NO, 160 ppmv SO<sub>2</sub>, SV = 6000 h<sup>-1</sup>, T = 440-460°C). The catalyst is able to withstand sulphur: no deactivation was detected over a period of 550 hours under the above conditions.

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CLAIMS

1. Method for the selective catalytic reduction of nitrous oxide ( $N_2O$ ) in the presence of a solid catalyst, with the addition of a saturated hydrocarbon as reducing agent, characterized in that the catalyst used is a promoted iron-containing zeolite.
2. Method according to Claim 1, characterized in that the catalyst is promoted with a precious metal.
3. Method according to Claims 1 and 2, characterized in that the precious metal comprises ruthenium, rhodium, palladium, gold or platinum, or a combination of two or more of these metals.
4. Method according to Claim 1, characterized in that the reducing agent used is natural gas or methane ( $CH_4$ ) or propane ( $C_3H_8$ ) or LPG ( $C_3H_8/C_4H_{10}$ ), or a combination thereof.
5. Method according to Claims 1 and 4, characterized in that the concentration of the reducing agent is set in such a way that the hydrocarbon/ $N_2O$  molar ratio is in the range from 0.2 to 20, preferably in the range from 0.5 to 5.
6. Method according to Claim 1, characterized in that the reduction takes place at an inlet temperature of less than  $400^\circ C$ .
7. Method according to Claim 1, characterized in that the emission of carbonmonoxide (CO) and of hydrocarbon is in each case separately less than 100 ppmv.
8. Method according to Claim 1, characterized in that the process pressure at which the reduction takes place is between 1 and 50 bar absolute.



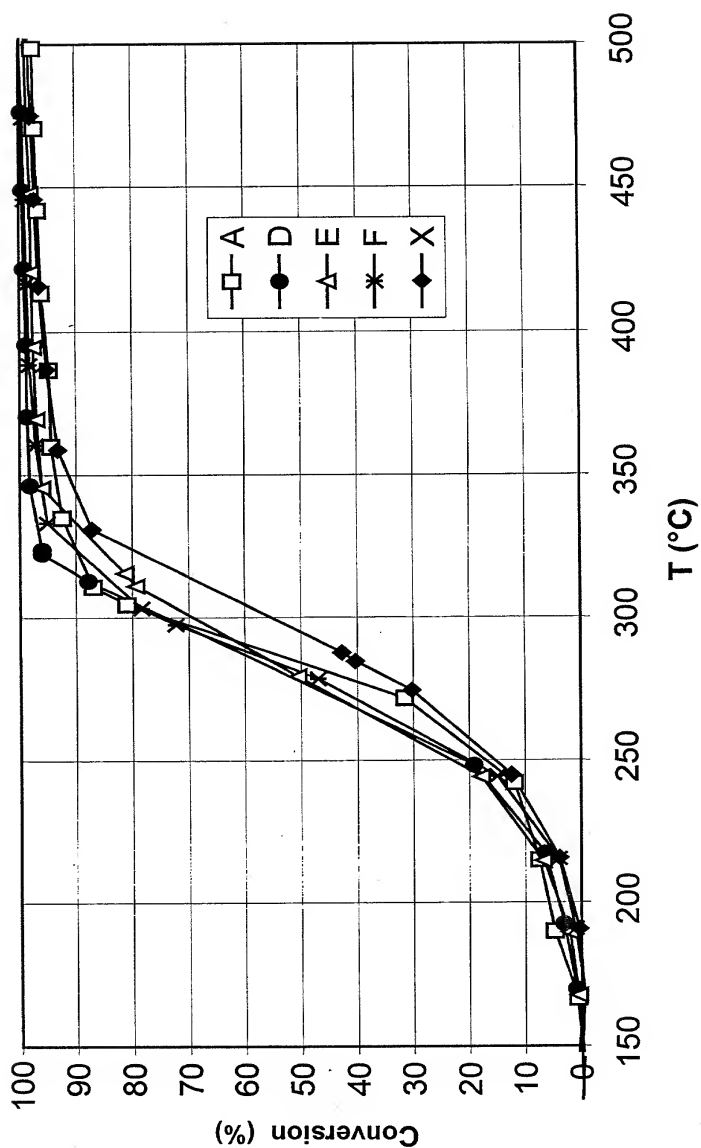


Fig. 1

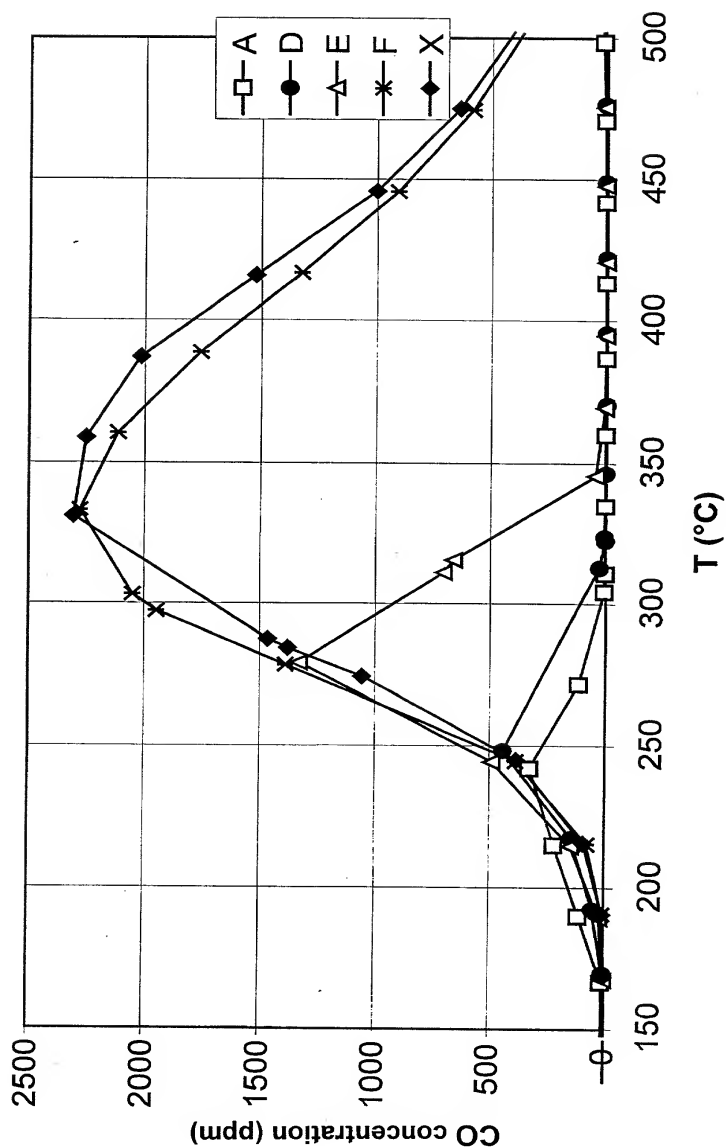


Fig. 2

# COMBINED DECLARATION AND POWER OF ATTORNEY

(ORIGINAL DESIGN, NATIONAL STAGE OF PCT OR CIP APPLICATION)

As a below named inventor, I hereby declare that

My residence, post office address and citizenship are as stated below next to my name, I believe I am the original, first and sole inventor (if only one name is listed below) or an original, first and joint inventor (if plural names are listed below) of the subject matter which is claimed and for which a patent is sought on the invention entitled:

**Reduction in N<sub>2</sub>O emissions.**

the specification of which: (complete (a), (b) or (c) for type of application)

## REGULAR OR DESIGN APPLICATION

a. ☐ is attached hereto.

b. ☐ was filed on  
Serial No.

as Application  
and was amended on

(if applicable)

## PCT FILED APPLICATION ENTERING NATIONAL STAGE

c. ☒ was described and claimed in International application No. PCT/NL00/00626 ✓  
filed on 6 September 2000 ✓

and as amended on

(if any)

## ACKNOWLEDGEMENT OF REVIEW OF PAPERS AND DUTY OF CANDOR

I hereby state that I have reviewed and understand the contents of the above identified specification, including the claims, as amended by any amendment referred to above.

I acknowledge the duty to disclose information which is material to patentability as defined in Title 37, Code of Federal Regulations, paragraph 1.56(a).

In compliance with this duty there is attached an information  
disclosure statement 37 CFR 1.97

## PRIORITY CLAIM

I hereby claim foreign priority benefits under Title 35, United States Code paragraph 119 of any foreign application (s) for patent of inventor's certificate listed below and have also identified below any foreign application for patent of inventor's certificate having a filing date before that of the application on which priority is claimed.

- d. ☐ no such applications have been filed  
 e. ☒ such applications have been filed as follows

**EARLIEST FOREIGN APPLICATION(S), IF ANY FILED WITHIN 12 MONTHS  
 (6 MONTHS FOR DESIGN) PRIOR TO SAID APPLICATION**

Country	Application Number	Date of filing (day, month, year)	Date of Issue (day, month, year)	Priority claimed
the Netherlands	1012983 ✓	6 September 1999 ✓		Yes
the Netherlands	1013862 ✓	15 December 1999 ✓		Yes

**ALL FOREIGN APPLICATION(S), IF ANY FILED MORE THAN 12 MONTHS  
 (6 MONTHS FOR DESIGN) PRIOR TO SAID APPLICATION**

**CONTINUATION-IN-PART**

(Complete this part only if this is a continuation-in-part application)

I hereby declare claim the benefit under Title 35, United States code, paragraph 120 of any United States application(s) listed below and, insofar as the subject matter of each of the claim of this application is not disclosed in the prior United States application in the manner provided by the first paragraph of Title 35, United States Code, paragraph 112, I acknowledge the duty to disclose material information as defined in Title 37, Code of Federal Regulations, paragraph 1.56(a) which occurred between the filing date of the prior application and the national or PCT international filing date of this application:

(Application Serial No.) (Filing date) (Status) (patented, pending, abandoned)

(Application Serial No.) (Filing date) (Status) (patented, pending, abandoned)

**POWER OF ATTORNEY**

As a named inventor, I hereby appoint the following attorney(s) to prosecute this application and transact all business in the Patent and Trademark Office connected therewith: Robert J. PATCH, Reg. No. 17,355, Andrew J. PATCH, Reg. No. 32,925, Robert F. HARGEST, Reg. No. 25,590, Benoît CASTEL, Reg. No. 35,041, Eric Jensen, Reg. No. 37,855, and Thomas W. PERKINS, Reg. No. 33,027 and Roland E. Long, Jr. Reg. No. 41,949 c/o YOUNG & THOMPSON, Second Floor, 745 South 23rd Street, Arlington, Virginia 22202.

Address all telephone calls to Young & Thompson at 703/521-2297.



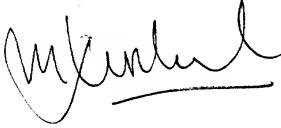
00466

PATENT, TRADEMARK OFFICE

I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that wilful false statements and the like so made are punishable by fine or imprisonment, or both under Section 1001 of Title 18 of the United States Code and that such wilful false statements may jeopardize the validity of the application or any patent issued thereon.

Full name of sole or first inventor: VERHAAK, Michael Johannes Franciscus Maria

Inventor's signature



Date 27/01/02

Country of Citizenship: the Netherlands ✓

Residence: ALKMAAR, the Netherlands NLX

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CHECK PROPER BOX(ES) FOR ANY ADDED PAGE(S) FORMING A PART OF THIS DECLARATION